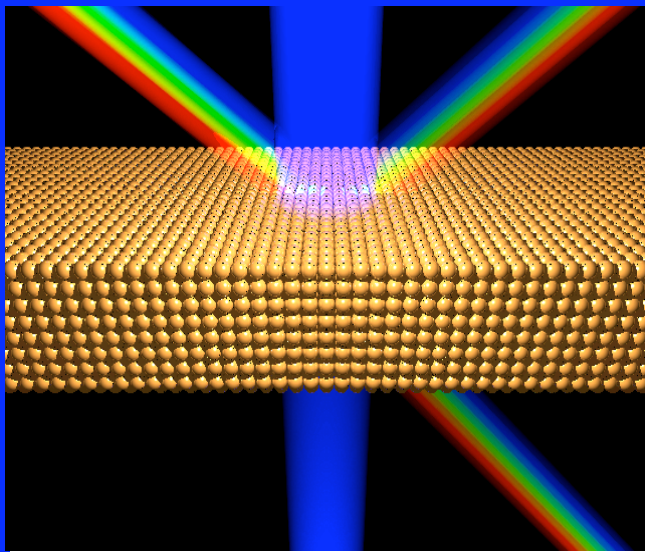


Isochoric Laser Heating for the study of Warm Dense Matter

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Outline



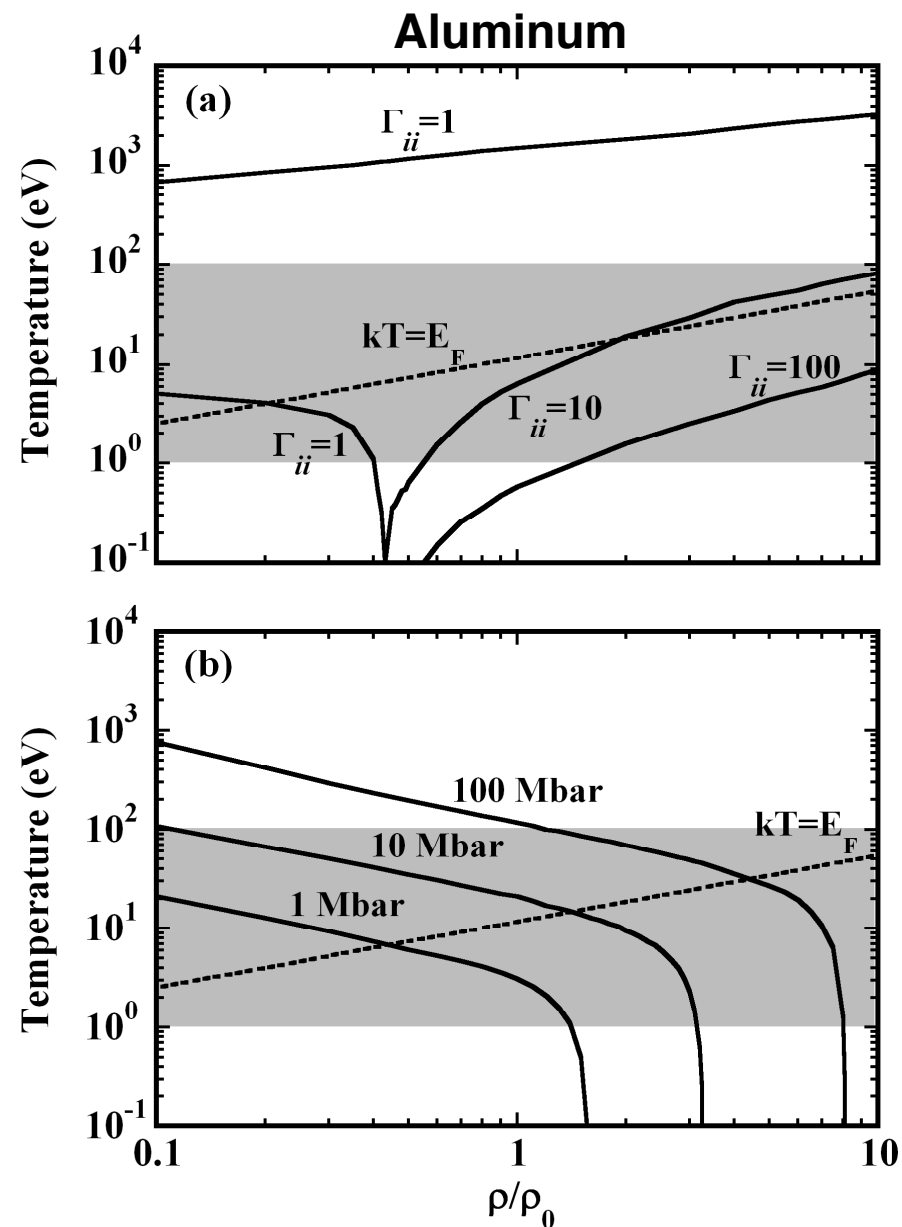
- **Introduction**
 - What is *Warm Dense Matter*?
 - *Idealized Slab Plasma & Isochoric Laser Heating*
- **Physics under non-equilibrium, extreme conditions**
 - Electrical conductivities
 - Lattice stability
 - Band structure and electron density of state

These are university-scale experiments

What is Warm Dense Matter?



- WDM introduced in 2000, characterized by
 - $kT \sim E_{\text{Fermi}}$
 - $\Gamma_{ii} = [P.E./K.E.]_{\text{ions}} > 1$
- Many-body, disordered system
 - Partial electron degeneracy
 - Excited electronic states
 - Pressure ionization
 - Strong ion-ion correlation
- High-pressure system
 - WDM is also HED Matter (>1 Mbar or 10^{11} J/m³)
 - Inertial confinement only
 - Rapid expansion



Warm Dense Matter is both fundamentally important and of broad relevance

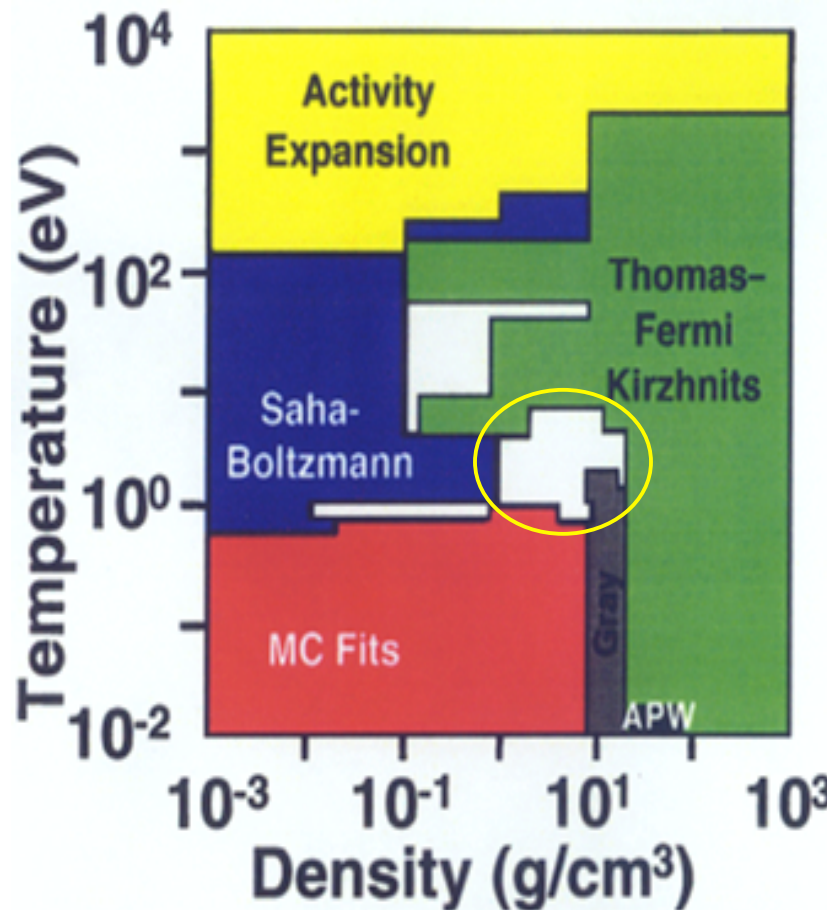


- **As finite-temperature condensed matter or strongly-coupled degenerate plasma, WDM is the basis for understanding the convergence of condensed matter and plasma science**
- **WDM finds applications in many disciplines**
 - **High Energy Density physics**
 - **Inertial Confinement Fusion**
 - **Shock physics**
 - **Material science**
 - **Planetary science**

WDM is an uncharted frontier as readily seen from the widely use EOS table - Sesame



Sesame EOS for copper
[K. Trainor, JAP (1983)]



APW - Electron band theory at 0K

GRAY - Semi-empirical Gruneisen-Debye theory for solid-melt-liquid

MC - Soft Sphere (Expanded liquid, vapor)

OCCIPITAL - Saha ionization equilibrium

TFNUC - Thomas-Fermi-Kirzhnits theory with semi-empirical nuclear corrections

ACTEX - Perturbation theory for high temperature ionization equilibrium

A critical void appears in the Warm Dense Matter regime

A major hurdle in WDM studies is the lack of single-state data



- **Laboratory WDM tends to be non-uniform due to hydrodynamic expansion at extreme pressure**
- **Properties measured on non-uniform or multi-state systems can only be compared with theory through code simulations that take into account gradient effects**

Unambiguous tests of theory requires

- Single-state physical data**
- Directly observed state parameters**

The concept of an *Idealized Slab Plasma* offers a means to achieve single-state measurements



- An *Idealized Slab Plasma* is a planar plasma that can be considered as a single uniform state in which any residual non-uniformities will impose negligible impact on the measurement of its uniform properties
- The state can be characterized from direct measurements such as mass density and energy density

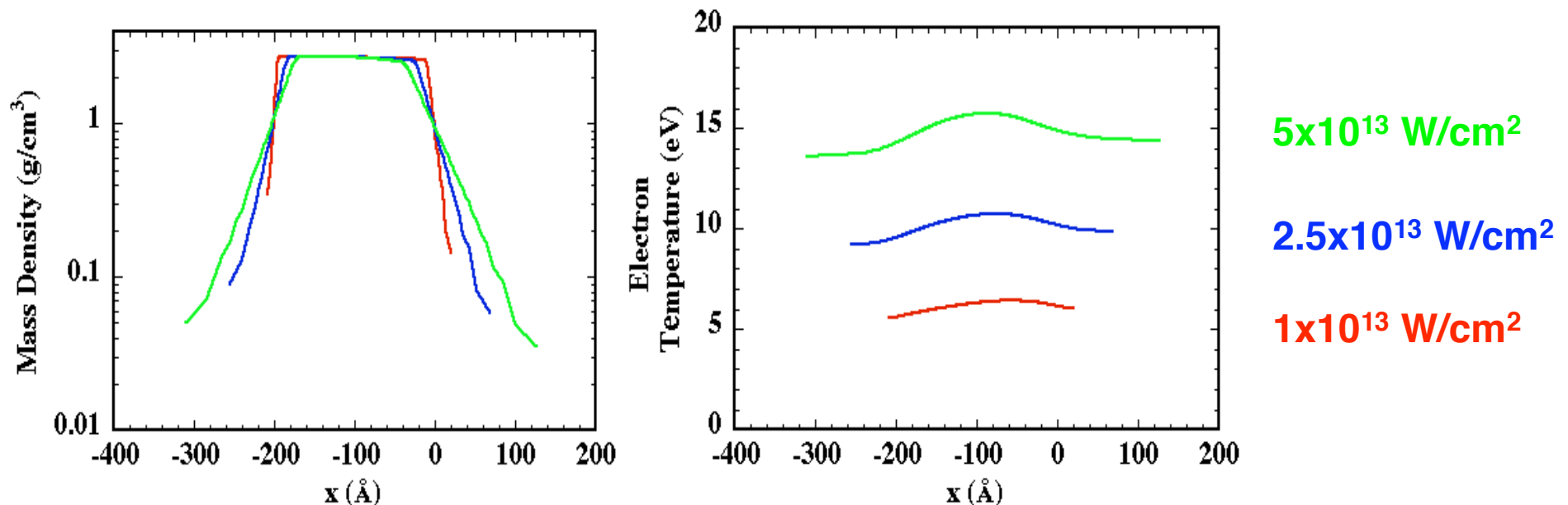
An approach to realize the *ISP* concept is *Isochoric Laser Heating* of a solid



- Laser heating in the *fs* time scale mitigates hydro expansion to yield **isochoric** condition
- Matching sample thickness to range of laser deposition or conduction scale length yields **isothermal** condition

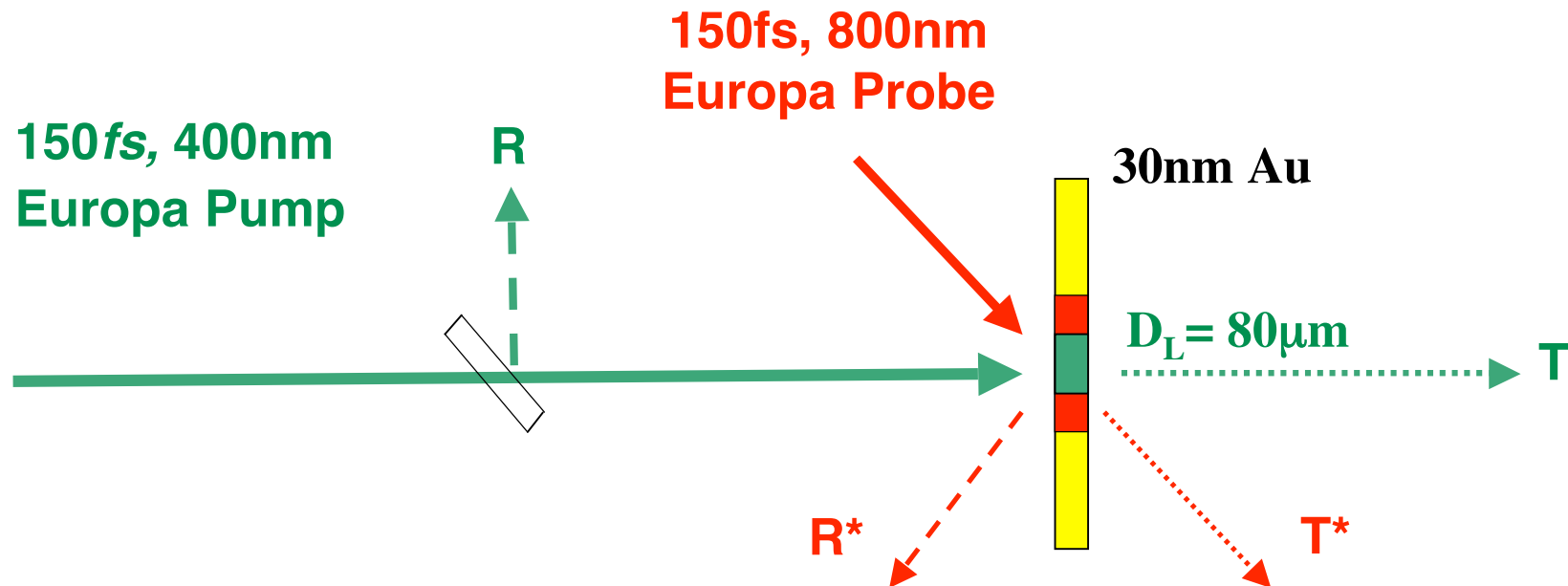
Forsman *et al.*, PRB 58, R1248 (1998)

20nm Al heated with a 100fs, 400nm laser



Isochoric heating is scalable to X-rays, electrons, protons or ions

The first *ILH* experiment is the measurement of electrical conductivity of warm dense Au



- Isothermal heating produced by laser skin-depth deposition and ballistic electron transport
- Isochoric condition maintained by material strength & inertia

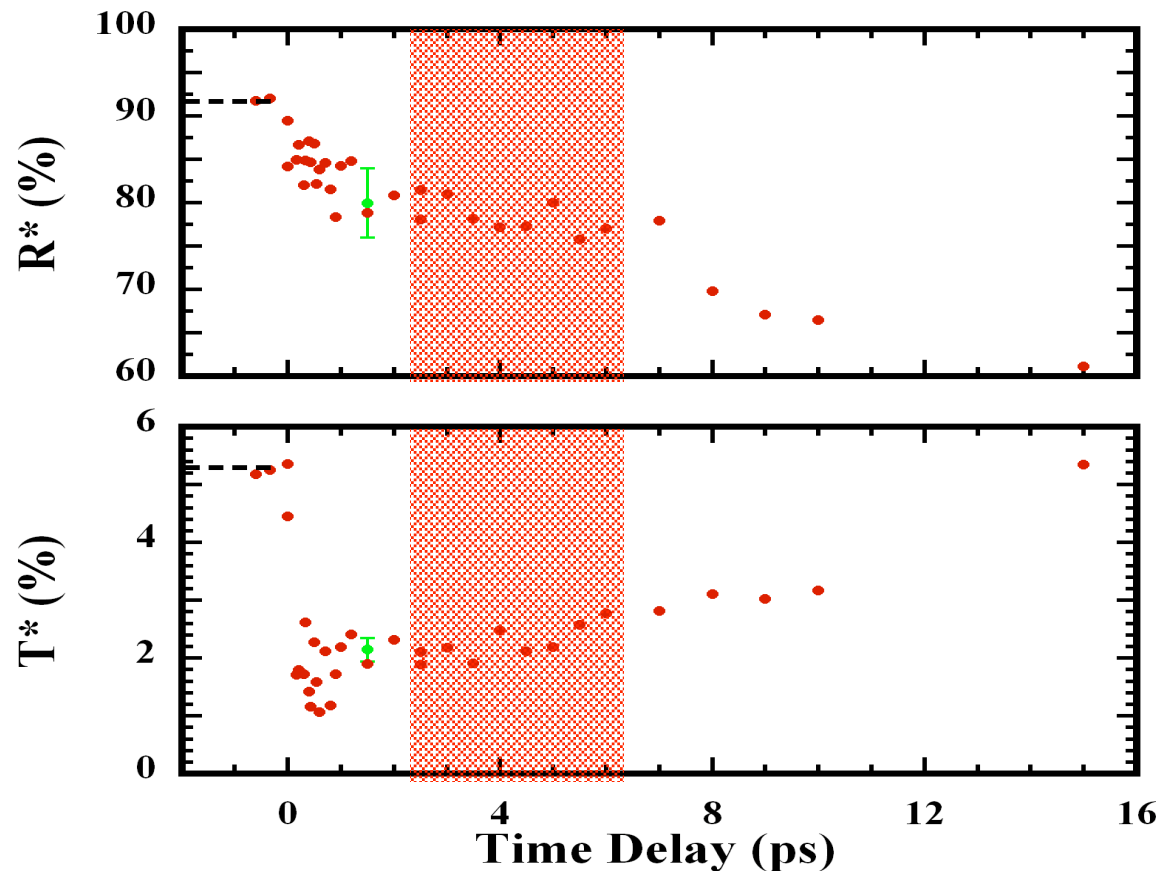
WDM state characterized by ρ_0 and $\Delta\epsilon$
– $\Delta\epsilon$ determined directly from $\{R, T\}$ of pump laser

Probe $\{R^*, T^*\}$ yields single-state data on $\sigma(\rho_0, \Delta\epsilon)$

Measurements of S-pol {R*, T*} reveal an interesting temporal behavior



- Three distinct stages are observed
 - An initial transient
 - **Quasi-steady state**
 - Hydrodynamic expansion



S-pol probe

$$\Delta\varepsilon = (3.5 \pm 1.0) \times 10^6 \text{ J/kg}$$

Similar behavior seen
with P-pol probe

Quasi-steady-state behavior is unexpected



- **Hydrodynamic simulations suggest disassembly of the foil in ~ 1 ps after heating when the lattice reaches melting temperature**
 - **Expansion gives rise to a plasma gradient on the surface of the foil; the gradient scale length will continue to increase with time**
 - **To maintain constant probe R^* and T^* , it would require the dielectric properties of the non-uniform system to evolve in a manner that precisely mitigates gradient effects at all times**

This is improbable

The problem of hydro code is the lack of solid state effects

Quasi-steady-state behavior has important consequences



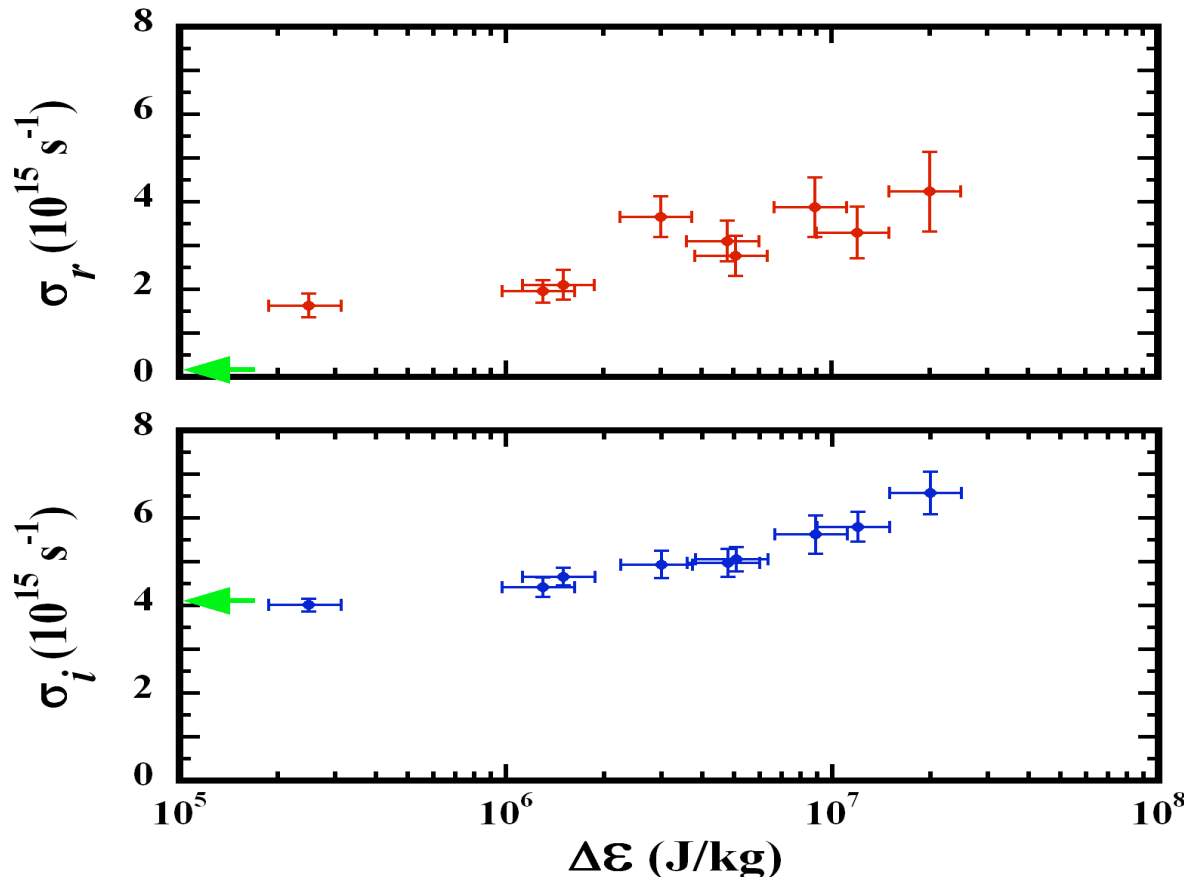
- It confirms the absence of significant hydrodynamic expansion, preserving the uniform, slab structure of the heated foil
- It yields an uniform state that is characterized by the direct observables of mass density ρ_o and excitation energy density $\Delta\varepsilon$

This ensures realization of the *Idealized Slab Plasma* concept in isochoric heating of a solid by *fs* laser

The quasi-steady state validates single-state measurement of AC conductivity



- Probe $\{R^*, T^*\}$ data for quasi-steady state used to solve Helmholtz eqs. for EM wave in a uniform dielectric slab
- This yields $\sigma_\omega(\rho_o, \Delta\epsilon)$ as direct benchmark for theory



Results obtained from
800nm, *S-pol* probe

We can learn more if we assume nearly free electron behavior

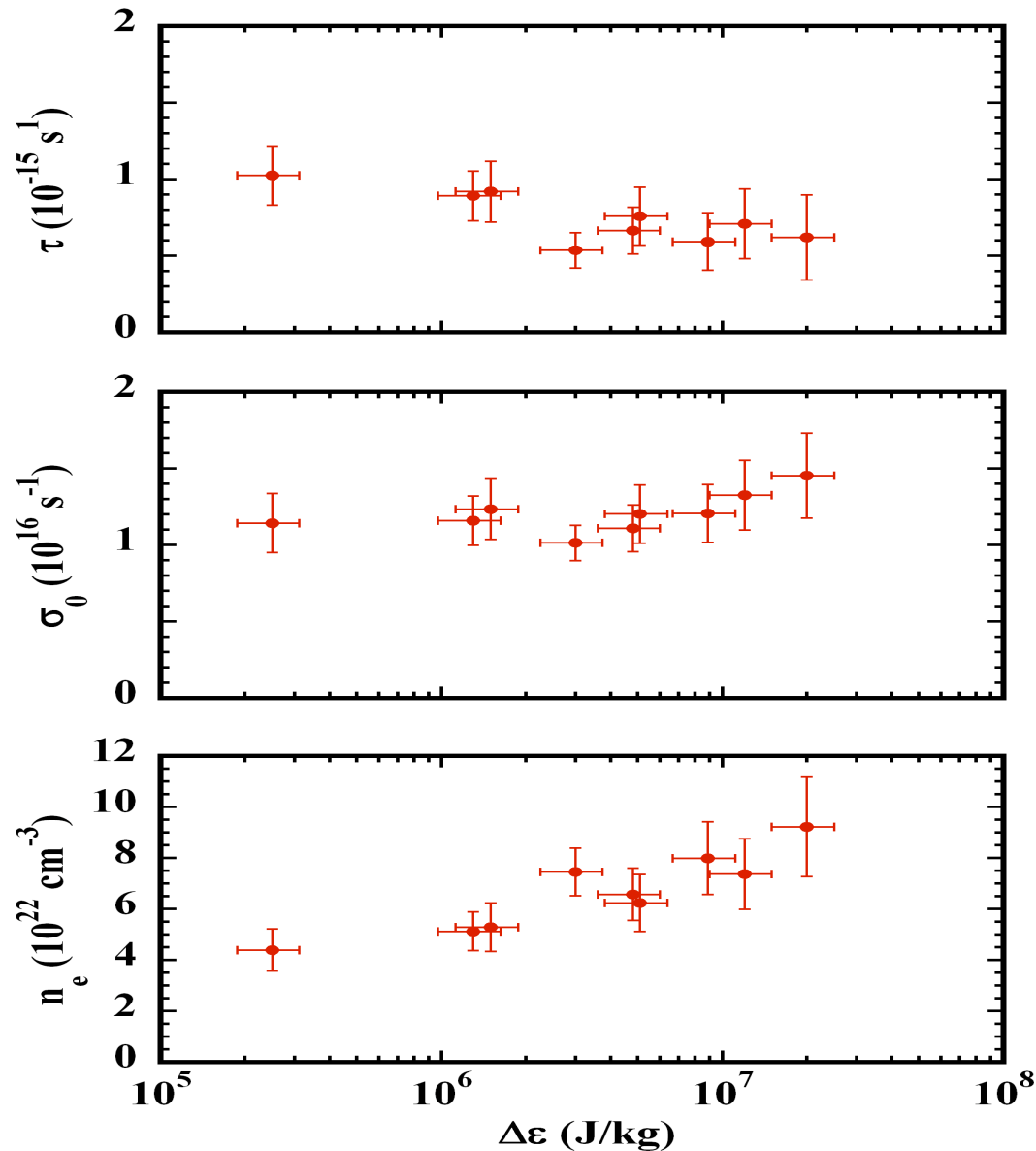


- **Nearly free electron behavior is expected**
 - Absence of interband transition at 800 nm
 - Conductivity effected by electrons near Fermi surface

- **Drude model:**
$$\sigma(\omega) = \sigma_r + i\sigma_i = \frac{\sigma_o}{1 + \omega^2\tau^2} (1 + i\omega\tau),$$

$$\tau = \frac{\sigma_i}{\sigma_r} \frac{1}{\omega}, \quad \sigma_o = \sigma_r (1 + \omega^2\tau^2), \quad n_e = \frac{m_e \sigma_o}{e^2 \tau}$$

This extends our single-state data
to include τ , σ_0 and $\langle Z \rangle$



At normal conditions:

$$\sigma_0 = 4.1 \times 10^{17} \text{ s}^{-1}$$

$$n_e = 3.8 \times 10^{22} \text{ cm}^{-3}$$

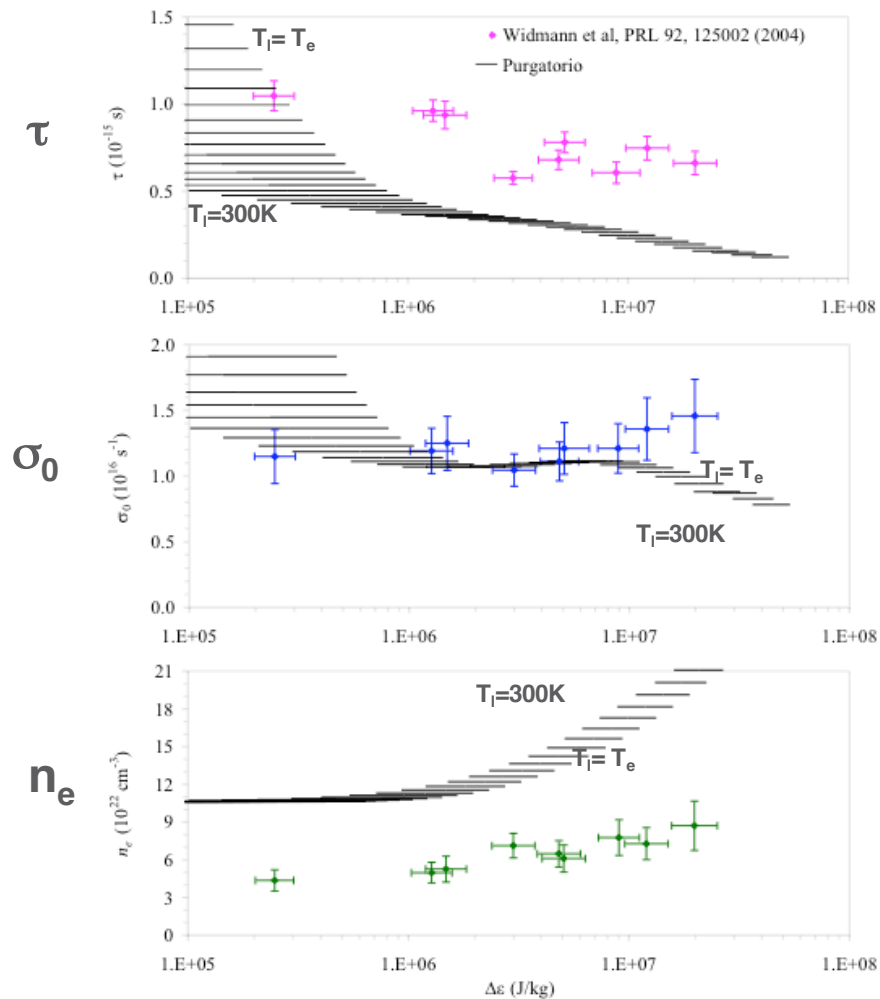
Widmann *et al.*,
PRL 92, 125002 (2004)

Drude behavior of σ at
800nm is subsequently
confirmed

The data provided the first benchmark of Purgatorio in the WDM regime



S. Hansen, B. Isaacs, V. Sonnad, P. Sterne, B. Wilson



- **Purgatorio Code**
 - Neutral-pseudo atom model
 - Dirac equ. for bound wave functions
 - Phase shifts by matching numerical wave functions to analytical forms at ion sphere radius
 - Bound & continuum electron density from Fermi distribution
 - Inelastic crystal structure factor [Baiko *et al.*, PRL 81, 5556 (1998)]
 - Electrical resistivity from extended Ziman formulation
- Agreement in σ_0 for $\Delta\epsilon < 10^7$ J/kg
- Discrepancy in τ , n_e
- Need for multi-parameter tests

What is the phase of the quasi-steady state?

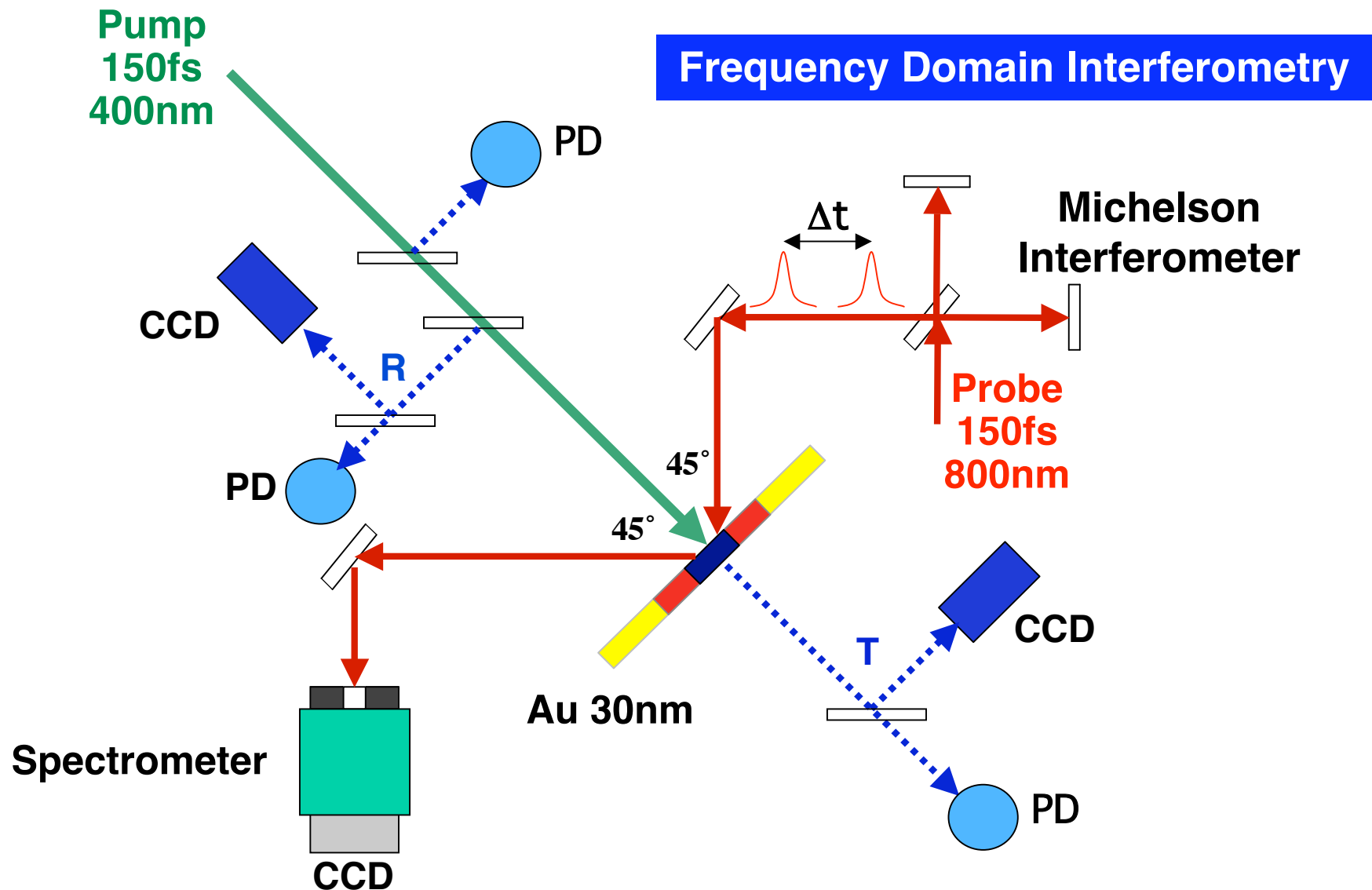


- Calculations of equation of state and transport properties require phase information, solid versus liquid, to determine the structure factor of the state
- The identity of the quasi-steady state is also key to understanding non-equilibrium phase transitions induced by ultrafast excitation

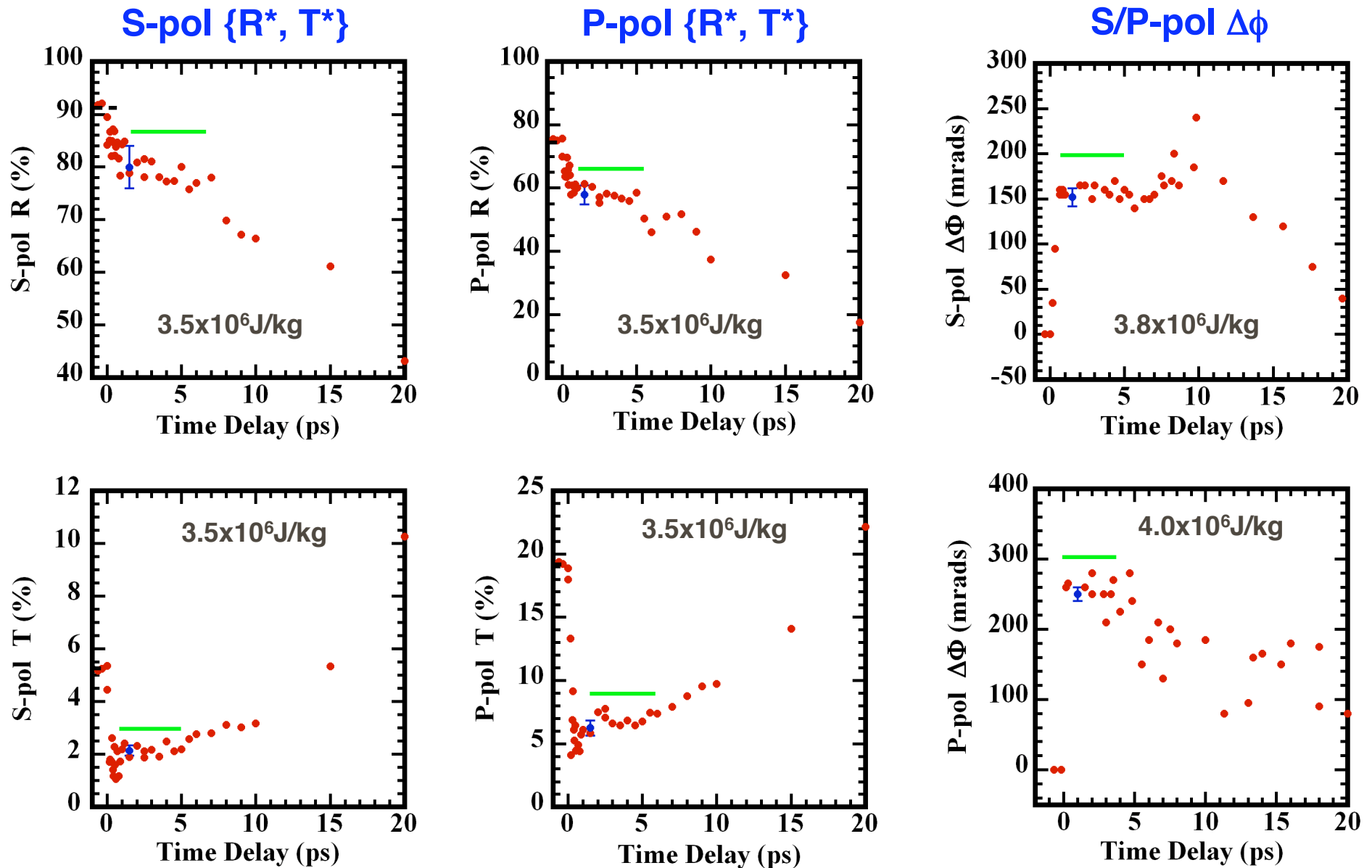
The immediate questions are

- If the lifetime of quasi-steady state is governed by stability of the lattice, is the limit set by a critical value of lattice energy density and can it be determined?
- Does the quasi-steady state retain any long or short range order?

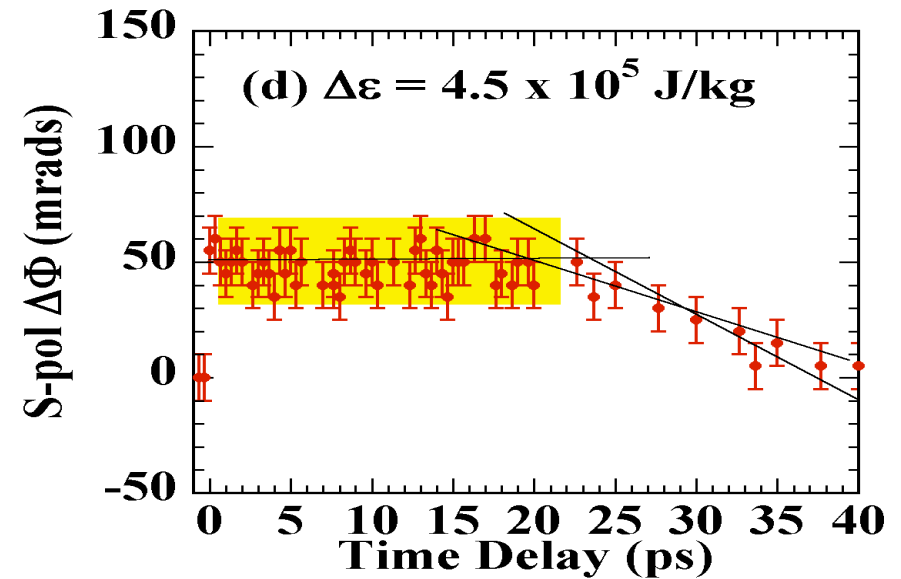
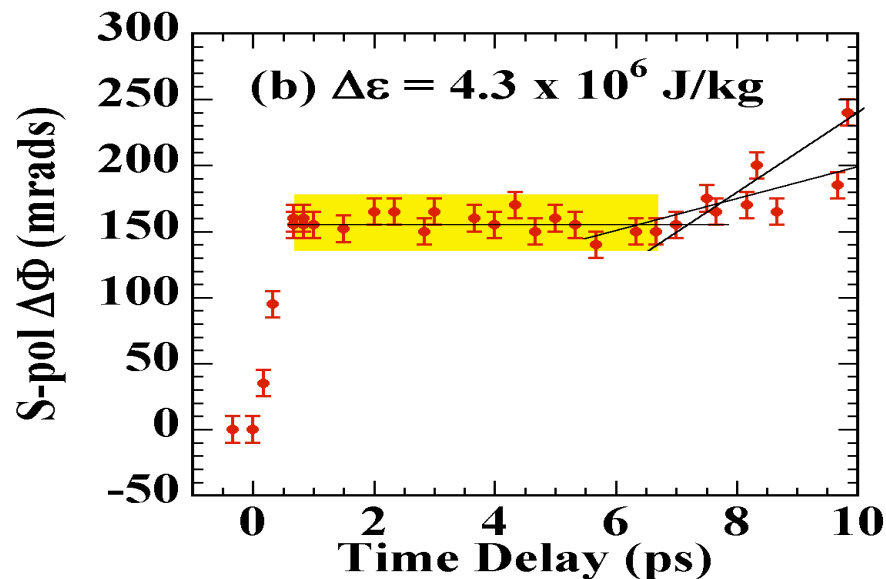
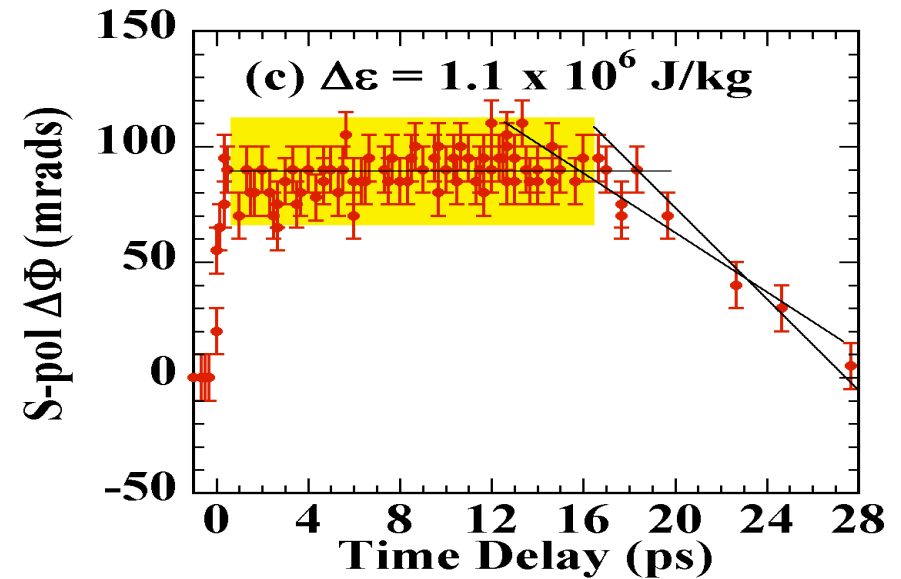
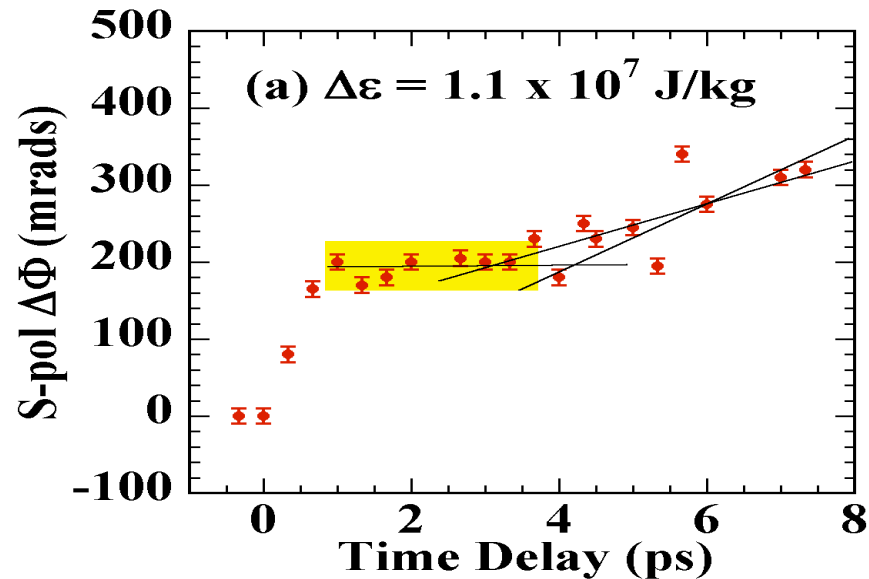
To determine lifetime of quasi-steady state, we probe hydro expansion with FDI



Quasi-steady state is confirmed in six different measurements



To quantify quasi-steady state duration,
we use an extensive set of S-pol FDI data



What are the processes governing solid-plasma transition in the heated foil?



- Laser heating of *s/p* electrons and photo excitation of *d*-electrons
- Electron-hole recombination
- Electron-electron thermalization
- Escape of heated electrons forming a surface sheath; sheath thickness is limited by space charge field
- Lattice heating effected by electron-phonon coupling
- Melting of the lattice
 - Ultrafast, non-thermal melting?
 - Thermal melting to meta-stable superheated liquid?
 - Superheated solid?
- Disassembly of the superheated state into a plasma

To describe lattice heating, we use a modified Two-Temperature Model



TTM:

$$C_e(T_e) \frac{dT_e(t)}{dt} = -g \left[T_e(t) - \varepsilon_l(t) \frac{\rho_{Au}}{C_l} \right] + S(t)$$
$$\rho_{Au} \frac{d(\varepsilon_l(t))}{dt} = g \left[T_e(t) - \varepsilon_l(t) \frac{\rho_{Au}}{C_l} \right], \quad \varepsilon_l(t) = \frac{C_l T_l(t)}{\rho_{Au}}$$

Electron-phonon coupling: $g = (2.2 \pm 0.3) \times 10^{16} \text{ W/m}^3 \cdot \text{K}^*$

Heat capacities: $C_e(T_e) = \frac{\partial U_e(T_e)}{\partial T_e}, \quad C_l = 2.5 \times 10^6 \text{ J/m}^3 \cdot \text{K}^\dagger$

Laser energy deposition: $S(t) = \frac{\Delta \varepsilon \rho_{Au}}{\tau_P \sqrt{\pi}} \exp\left(-\frac{t^2}{\tau_P^2}\right)$

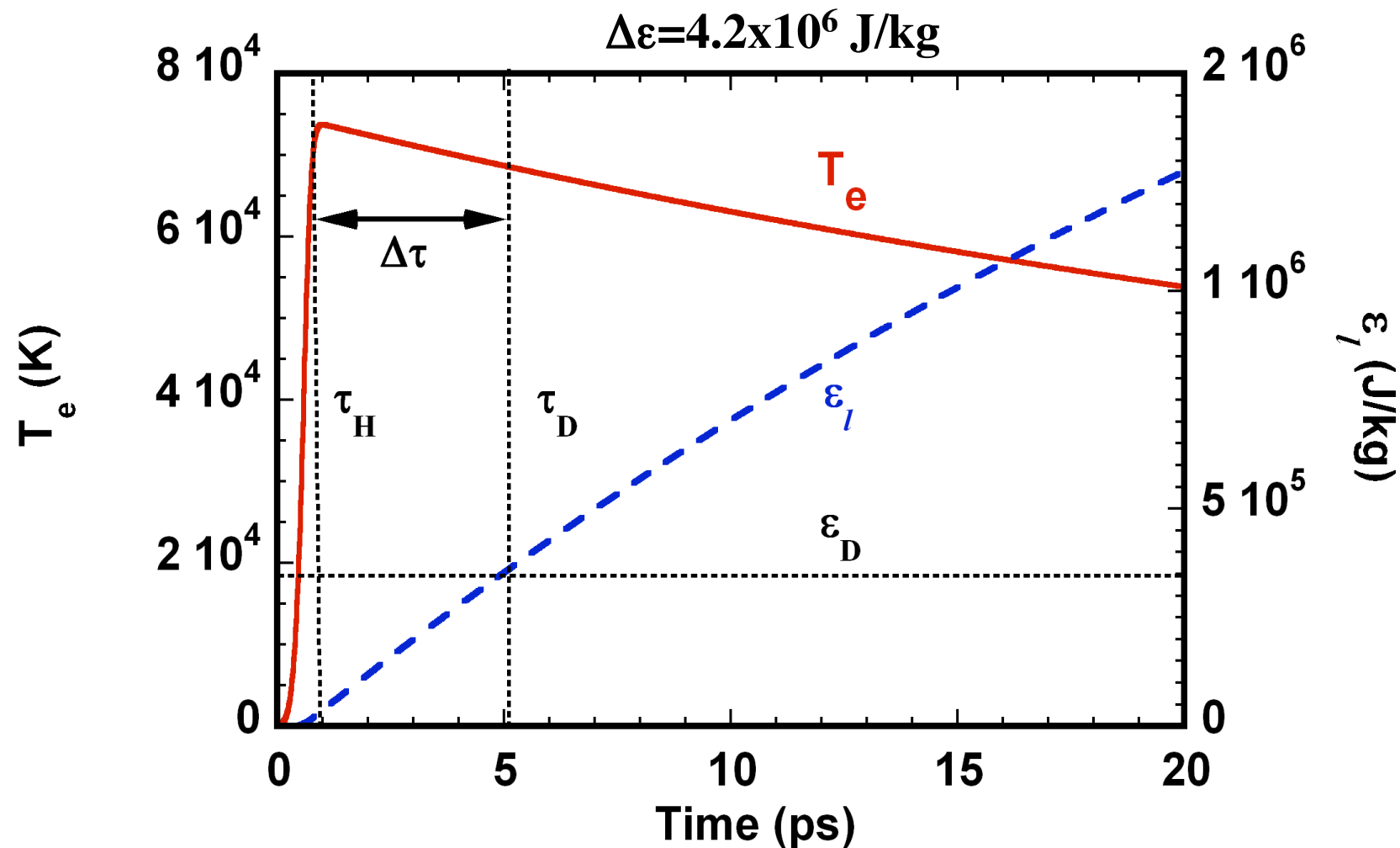
*Hohlfeld *et al.* Chem. Phys. 251, 237 (2000)

†Maxmillian's Chemical and Physical Data, Maxmillian Press, London, 1992

We postulate that disassembly is a rate-independent critical phenomenon



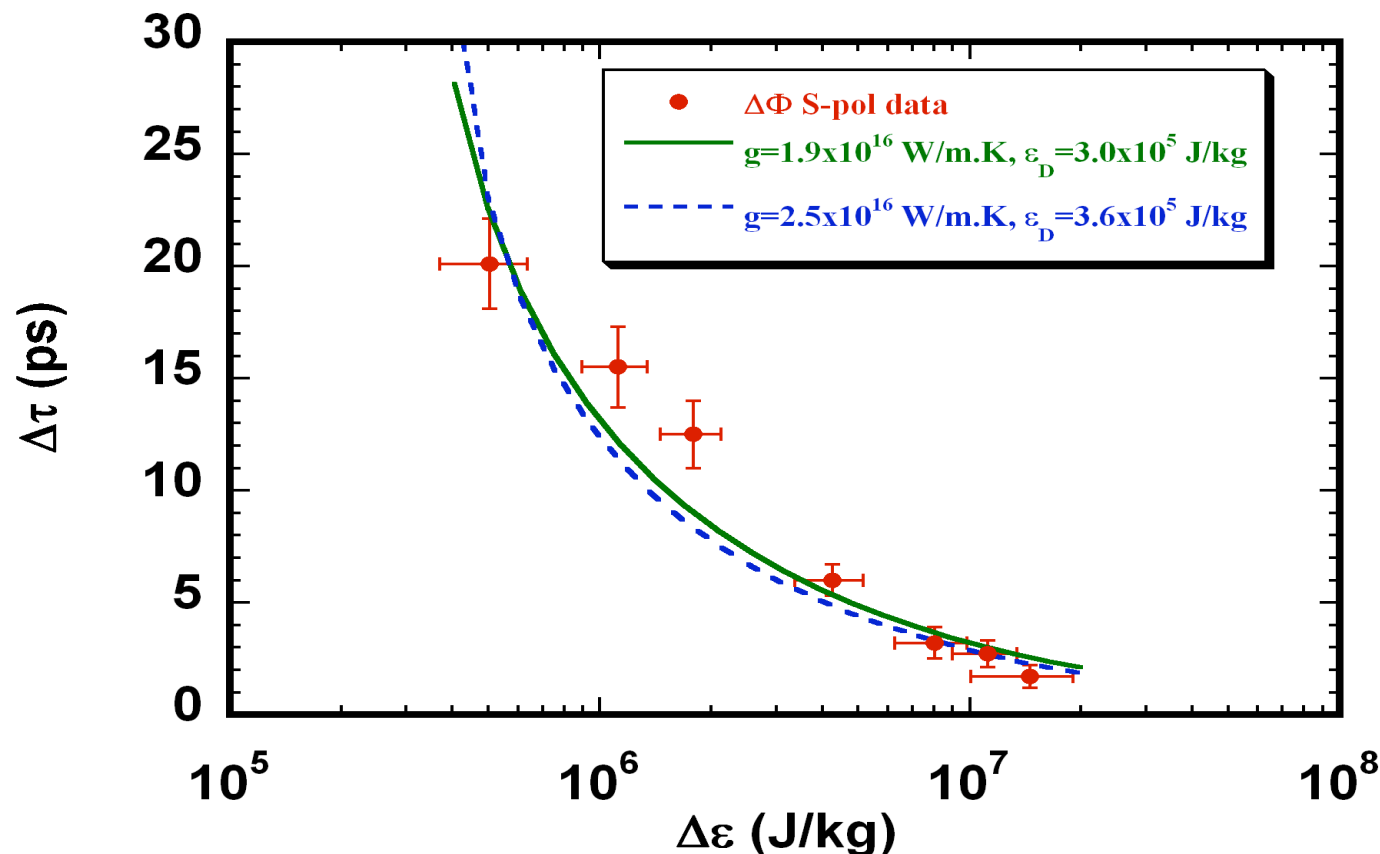
- Quasi-steady-state duration $\Delta\tau$ is determined by a critical value ε_D independent of heating rate (or $\Delta\varepsilon$)



The heating-disassembly model shows good agreement with observation



- This yielded the first measurement of the critical lattice energy $\varepsilon_D = (3.3 \pm 0.3) \times 10^5$ J/kg for solid-plasma transition under ultrarfast laser excitation

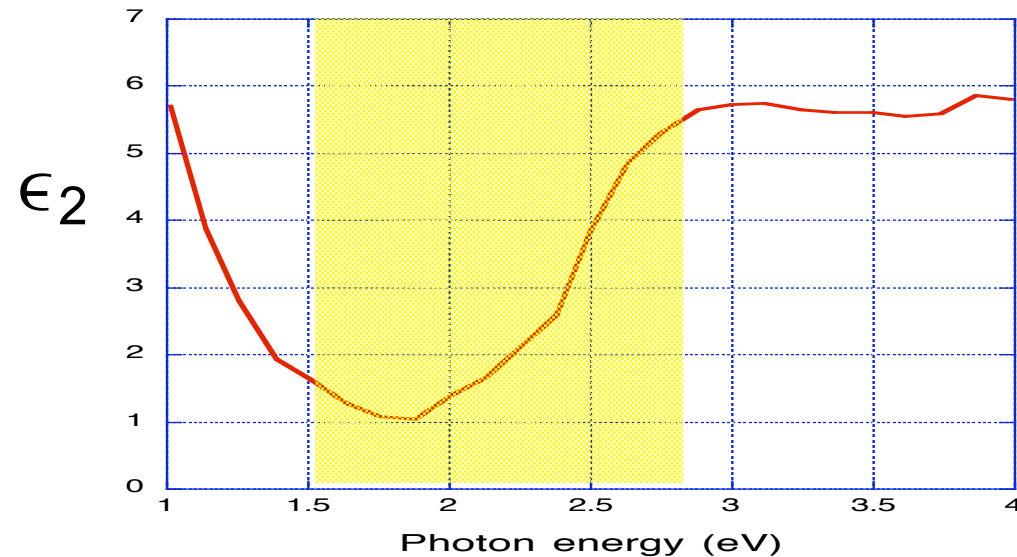


Ao *et al.*, Phys. Rev. Lett. 96, 055001(2006)

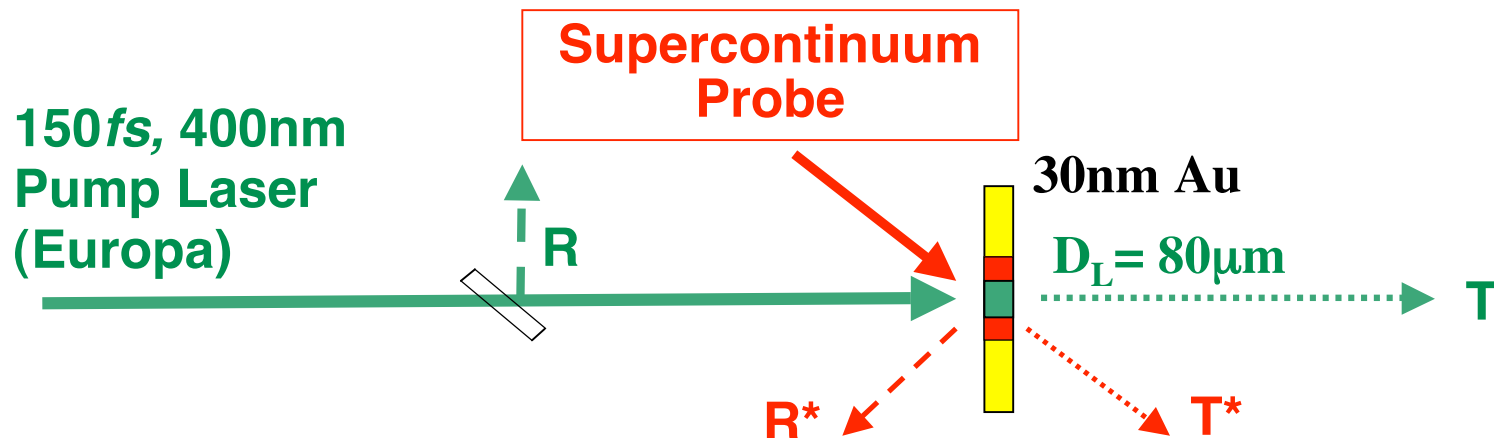
To probe long/short range order in quasi-steady state, we use broadband dielectric function



- For Au, intra & inter-band transitions in 450-800nm of $\epsilon(h\nu)$



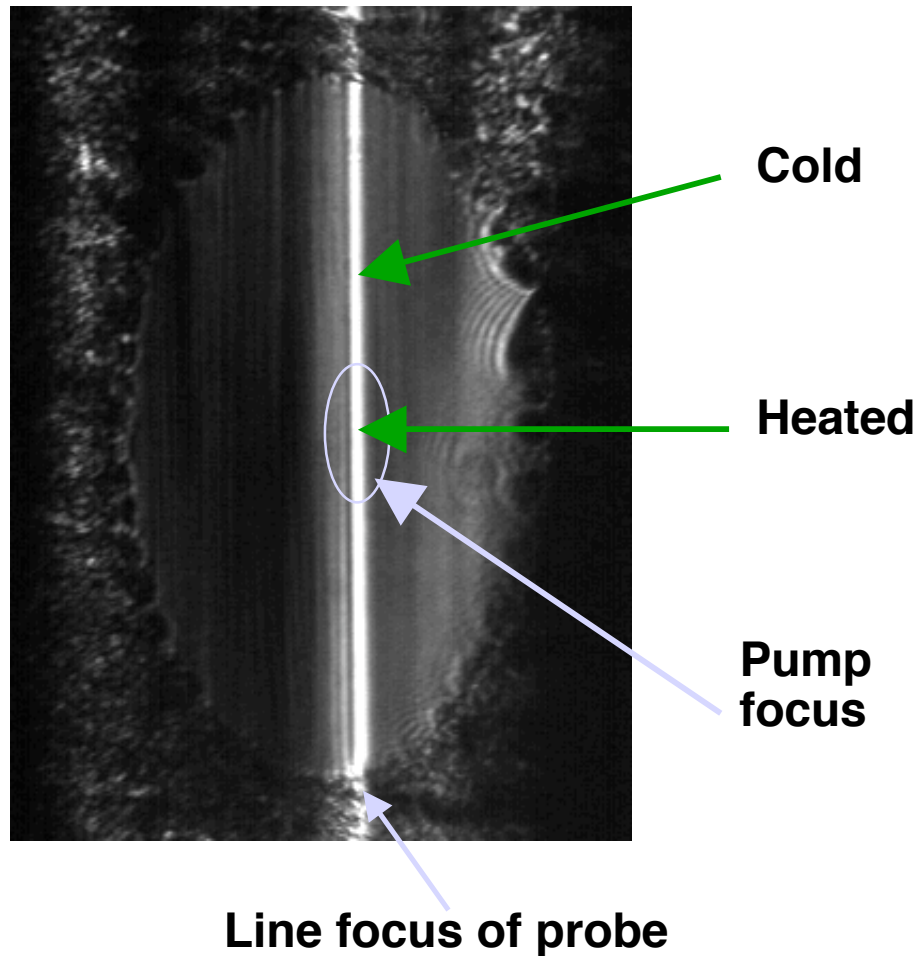
- $\epsilon(h\nu)$ determined from $\{R^*, T^*\}$ of supercontinuum probe



Probe $\{R^*, T^*\}$ measured with *in-situ* calibration

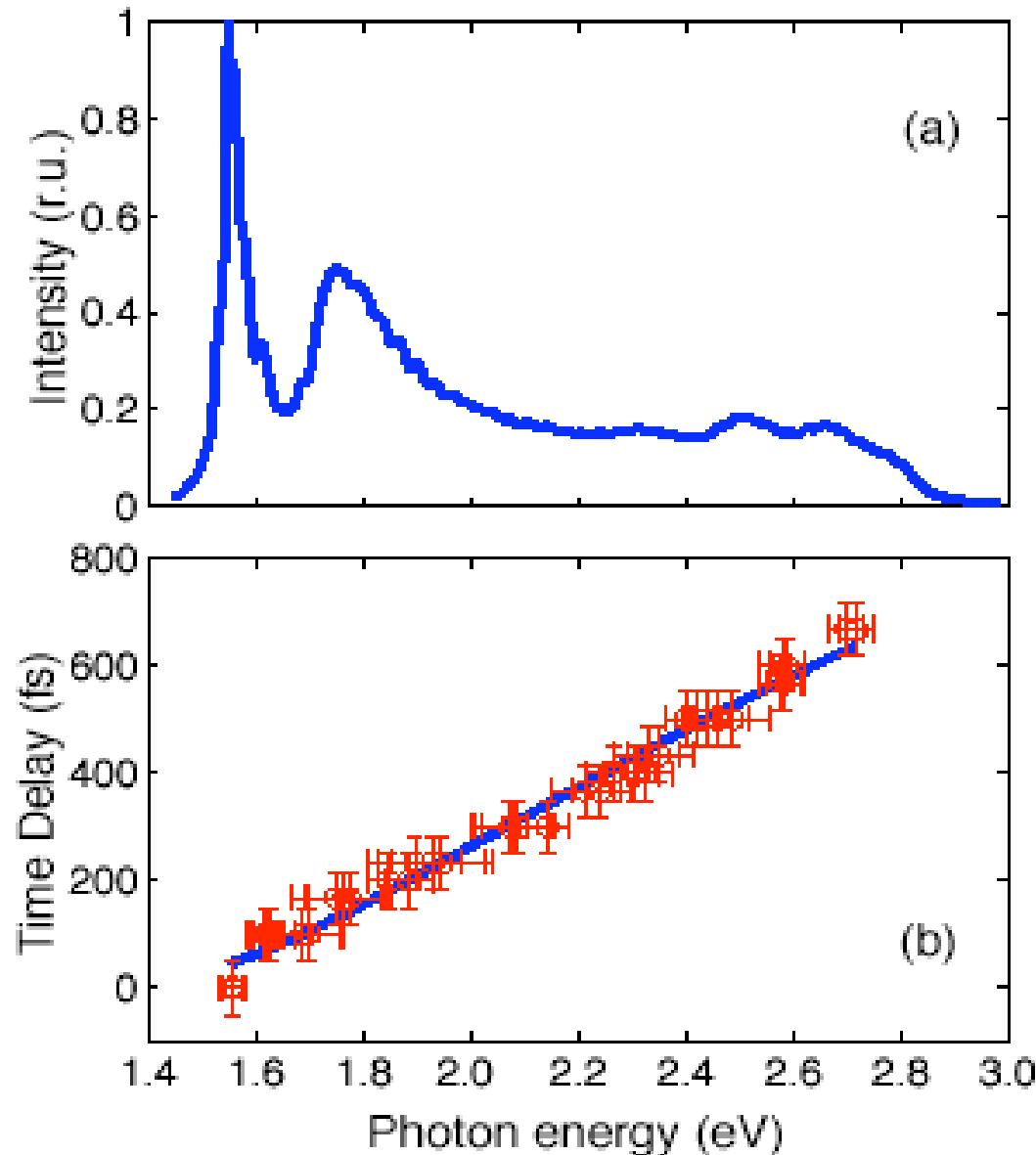


Reflectivity image of Au foil at 45°



- 180fs, 800nm laser is focused onto CaF_2 to generate a 450-800nm supercontinuum probe
- Probe illuminates nanofoil at 45°-incidence in $30\mu\text{m} \times 600\mu\text{m}$ line focus, covering both heated and unheated regions
- *In-situ* calibration eliminates the need for
 - Absolute intensity calibration
 - Measurement of shot-to-shot variation in probe intensity

Frequency chirp in supercontinuum is measured using Kerr optical gate

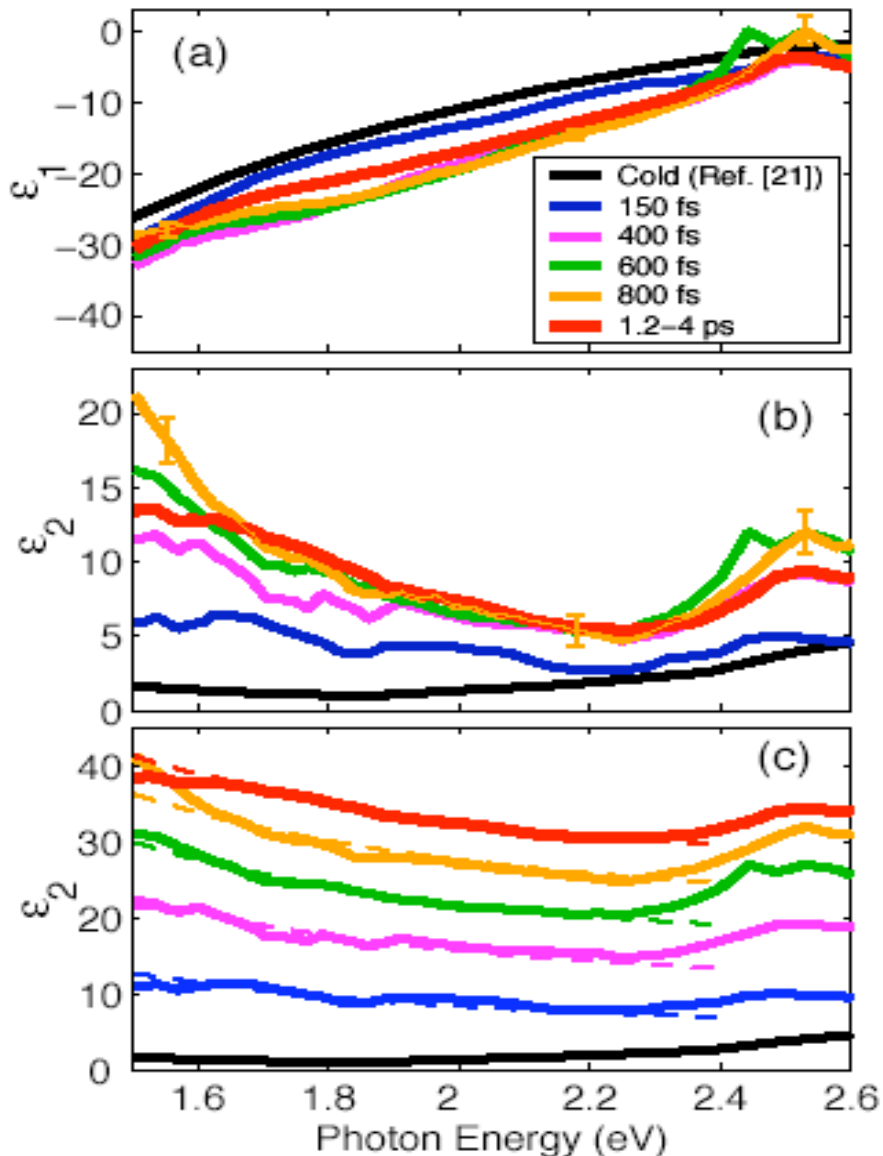


- Supercontinuum provides spectral measurements from 450-800 nm
- Frequency chirp gives rise to time-encoded spectrum
 - To remove effect of chirp in measurements
 - Bin spectral data in 10nm intervals
 - Apply temporal shifts using chirp data

Temporal evolution of $\epsilon(h\nu)$ of Au at 2.9×10^6 J/kg



Data corrected for frequency chirp

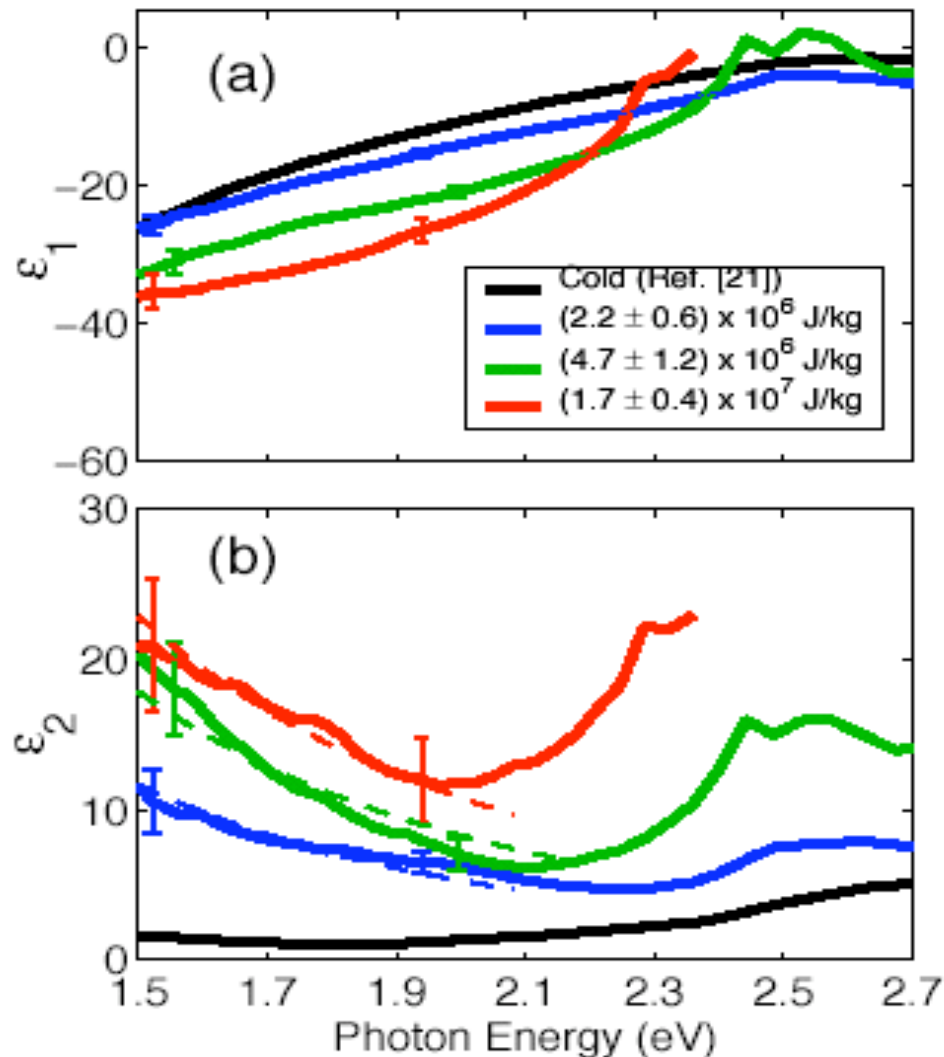


- Quasi-steady-state behavior seen in 1.2-4 ps consistent with earlier finding [Ao *et al.*, PRL 2006]
- $\epsilon_1(h\nu)$ relatively featureless
- $\epsilon_2(h\nu)$ shows distinct components
 - Intraband transitions below 2.3 eV
 - Enhancement in transitions
 - Overshoot at 1.55 eV similar to previous observation
 - Drude behavior
 - Interband transitions above 2.3 eV
 - Enhancement in transitions

Dependence of $\epsilon(h\nu)$ of Au on excitation energy density $\Delta\epsilon$

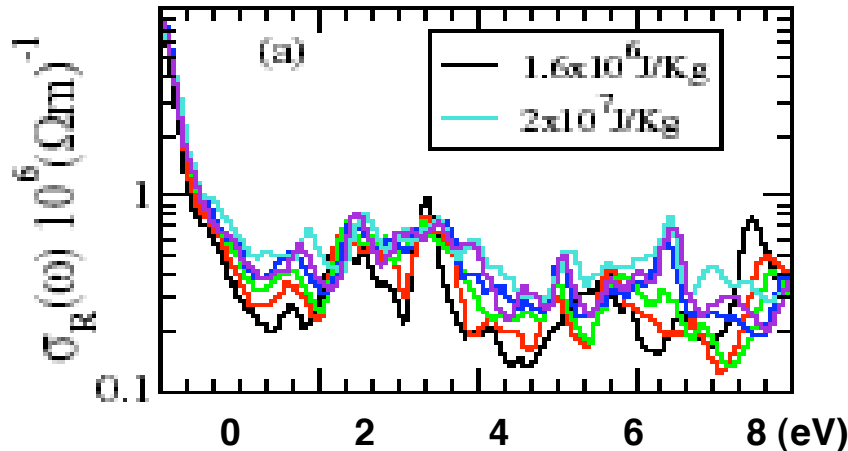


Probe delay varies from 1.4ps
@1.55eV to 2ps @2.6eV

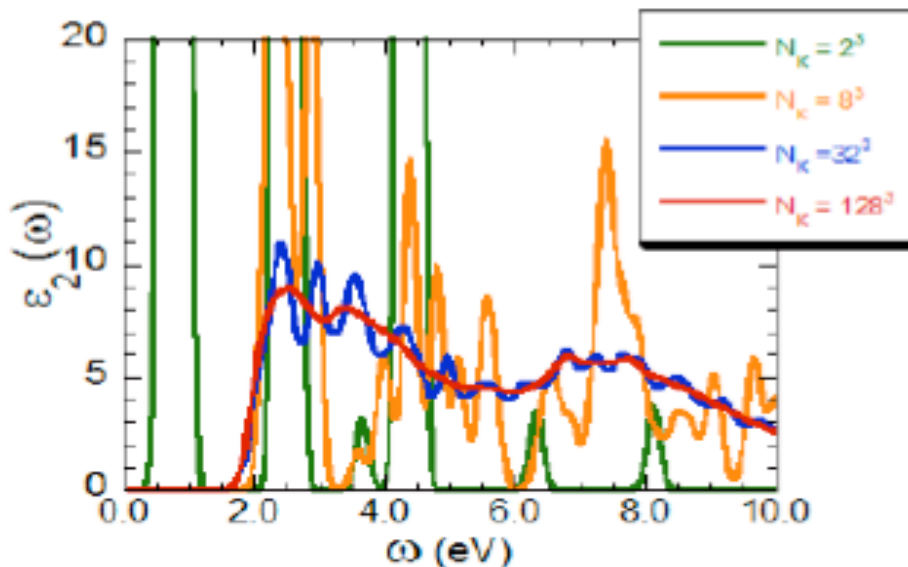


- For $\Delta\epsilon$ of 2.6×10^6 , 4.7×10^6 J/kg
 - The 1.4-2 ps probe delay falls within the quasi-steady-state
- For $\Delta\epsilon$ of 1.7×10^7 J/kg
 - Disassembly occurs at 2.38 eV for a probe delay of 1.9 ps, consistent with previous data
- Intra band transitions
 - Enhancement with $\Delta\epsilon$
 - Drude behavior
- Inter band transitions
 - Enhancement with $\Delta\epsilon$
 - Increasing red shift with $\Delta\epsilon$

Drude behavior in intra band transitions points to discrepancy in $\sigma(h\nu)$ calculation



- Spectral structures in $\sigma(\omega)$ above 1.3 eV were reported [Mazevet *et al.*, PRL 2005]
 - Sampling of Brillouin Zones over only $\sim 8^3$ k -points



- Limited BZ sampling can lead to spurious spectral structures [T. Ogitsu & E. Schwegler]
 - fcc Au at 0 K
 - Convergence is reached with 128^3 k -points

ϵ_2 data in disagreement with calculations lacking treatment of non-adiabatic effects of electron-phonon coupling

The prominence of inter band transitions raises many interesting questions



- **Persistence of d -band in the quasi-steady state**
 - If d -band is the result of long range order, this would be first evidence of the quasi-steady state being a superheated solid
- **Red shift can be due to temperature-induced changes in the energy distribution of the electrons**
- **Enhancement is likely a non-equilibrium effect**
 - Equilibrium calculations for Al shows disappearance of interband transitions at melting (Benedict *et al.*, PRB 2005)
 - Photoemission spectroscopy on fs-laser excited Au at $300\mu\text{J}/\text{cm}^2$ shows residual non-thermal electron distribution after 670fs (Fann *et al.*, PRB 1992)

Ping *et al.*, Phys. Rev. Lett. 96, 255003 (2006)

Summary



- The *Idealized Slab Plasma* Concept has been realized in *Isochoric Laser Heating*
- This has become a unique platform for the study of non-equilibrium, high-energy-density Warm Dense Matter free from gradient effects
 - Electrical conductivity
 - Lattice energy density for solid-plasma transition
 - Persistence of band structure in quasi-steady state with non-equilibrium electron DOS

Warm Dense Matter an emerging frontier in plasma & CM science

- 2000, 2002, 2005, 2007, 2009 International WDM Workshop
- 2002 LLNL Workshop on Extreme States of Material: WDM to NIF
- 2002 US-France Workshop on WDM
- 2003 CECAM Workshop on QMD Approaches of WDM
- 2006 Accelerator-Driven WDM Workshop
- 2006 Lance Dynamic Experiment Facility Workshop